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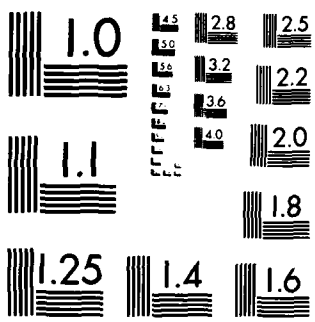

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Edward L. Ginzton Laboratory
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Stanford, California

LASER PHYSICS AND LASER TECHNIQUES

Final Scientific Report
for
Air Force Office of Scientific Research
Contract No. AFOSR F49620-82-K-0015
for the period
1 February 1982 — 31 January 1984

Principal Investigator:
Professor A.E. Siegman

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Quantum Wells	Nondegenerate Four-Wave Mixing	Colliding Pulse
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Surface Ripples	Thermal Gratings	Nonlinear Optics
Laser Damage	Photoacoustic Spectroscopy	

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

We have performed a two-year program of theoretical and experimental research on laser physics and laser techniques. We have observed time-resolved photoluminescence in GaAs quantum wells and have found it to obey a multicomponent decay law. Excite-probe experiments on semiconductors has resulted in a new technique for observing dense electron-hole plasmas just below melting. We have also studied and solved the electromagnetics of the formation of surface ripples under single-beam illumination. Using a novel three-laser frequency-domain optical technique, we have measured excited-state decays and orientational

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relaxation with subpicosecond resolution. In addition, we have theoretically analyzed phase conjugate resonators and thermal grating formation. Finally, we performed time-resolved photoacoustic spectroscopy with subnanosecond resolution using a novel elliptical cell, and also performed colliding-pulse mode-locking experiments in an antiresonant-ring arrangement.

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I. INTRODUCTION

This is the Final Scientific Report on AFOSR Contract F49620-82-K-0015 for the period 1 February 1982 through 31 January 1984. This report summarizes research, journal publications, and conference presentations in the areas of picosecond optical pulses and picosecond spectroscopy; laser-induced surface transformations and spontaneous "ripples"; tunable-laser-induced-grating spectroscopy; and optical phase conjugation and phase-conjugate resonators. A complete list of AFOSR-sponsored publications and conference presentations is attached.

II. REVIEW OF ACCOMPLISHMENTS

A. Recombination Times Between Confined States in Quantum-well Semiconductor Structures

Superlattices[1,2] and quantum wells are a relatively new generation of electronic materials made possible by advances in the growth techniques of molecular beam epitaxy (MBE) and metal organic chemical vapor deposition (MOCVD). Layers of individual semiconductor materials in these wafers are so thin that electrons and holes are quantum-mechanically confined in them. The structures we are investigating utilize very thin $L_z < 100$Å low-bandgap GaAs wells between higher-bandgap $Al_{.3}Ga_{.7}As$ barriers. The electrons and holes must sit only at certain allowed energy levels in the GaAs conduction and valence-band wells. These energy levels in a single well can be calculated using the finite-square-potential-well model from basic quantum mechanics. Recombination occurs between confined states in the conduction and valence band and the photons produced in this recombination have greater energies than the recombination photons in bulk GaAs because in quantum wells the carriers are confined in levels above the bottoms of the wells (see Fig. 1).

Semiconductor coupled-mode stripe lasers fabricated with quantum-well active layers have produced cw outputs[3] greater than 1 Watt, while bistable optical effects in quantum-well structures have shown useful nonlinearities at powers as low at 80 mW. Quantum-mechanical confinement of carriers at interface states (a concept similar to square wells) had led to a 2 Gigabit per second electronic integrated circuit[4]. For these and other reasons, the practical interest in quantum-well structures is great.

Despite the great interest in quantum-well structures, both for their applications and also for their own inherent appeal, the carrier dynamics in these structures has not yet been extensively studied. We have been studying the photoluminescence time for recombination between electrons in the first confined state in the conduction band and holes in the first confined state in the valence band. First, an initial determination of photoluminescence time versus excitation energy density was made using equipment borrowed from the Stanford Structural Biology Department. This equipment was available for only a limited length of time, and the conditions

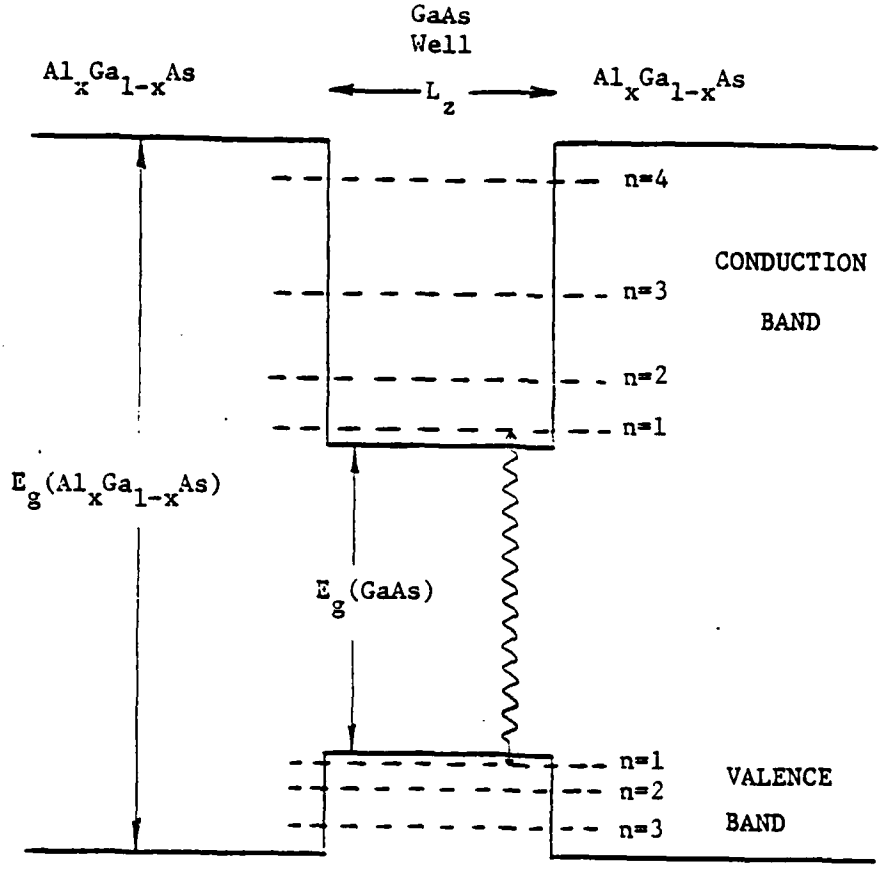

FIGURE 1

Single Quantum Well. The confined electron states are shown in the conduction-band well and the confined hole states are shown in the valence-band well. (There are two sets of valence states corresponding to heavy and light holes, but only one is shown for clarity). Carriers fall into the $n = 1$ levels and recombine between bands. The energy of the photons thus produced, $h\nu$, is greater than the bandgap of bulk GaAs, E_g(GaAs).

of the loan were not broad enough to allow operation at the long laser excitation wavelengths necessary to resolve certain questions which arose during these experiments. Hence, we have set up a second apparatus in our laboratory with the intention of measuring the room-temperature intrinsic photoluminescence time in a quantum well.

Previous time-resolved experiments on quantum wells have concerned excitonic effects. D.A.B. Miller, *et al*,[5] measured a $\tau = 21$ ns absorption recovery time for the $n = 1 \rightarrow 1$ heavy-hole transition in a $GaAs/Al_xGa_{1-x}As$ quantum well. This lifetime measurement was unfortunately affected by carriers diffusing out of the illuminated spot, and the recovery time was inferred using only two points and assuming a single-component decay. Even so, such a measurement cannot be used to predict photoluminescence times in a light-emitting device because absorption includes formation of an exciton (bound electron-hole pair). This exciton has a high probability of being ionized before photoluminescence occurs for the case of excitation at an energy reasonably larger than the quantum-well transition. Hence, our room-temperature photoluminescence measurements represent primarily free-carrier dynamics rather than exciton dynamics. Recombination times are expected to be different for exitons than for free carriers because the electron and hole are in close proximity in an exciton. E.O. Göbel, *et al.*[6] measured a 10 K photoluminescence time of $\leq$ 1 ns on this same quantum-well transition. However, at such a low temperature, there are few phonons present to ionize the excitons formed upon absorption so that recombination times reflect exciton dynamics. Masumoto and Shionoya[7] have also investigated exciton dynamics at liquid-helium temperatures. In both these experiments fluences at the high-photon-energy excitation wavelength were considerably greater than the absorption saturation intensity that Miller, *et al.*[5] measured on the $n = 1 \rightarrow 1$ exciton transition. Considering that not too much intraband relaxation takes place during the few ps excitation pulsewidths in these low-temperature experiments, it is probably safe to say that both of these experiments were demonstrating exciton-population saturation effects.

We have performed the majority of our experiments on photoluminescence at room temperature so that recombination would be dominated by free-carrier effects rather than excitonic effects. (We are planning 77 K experiments which will then allow us to compare free carrier-dominated and exciton-dominated lifetimes. In

addition, we will measure recombination time as a function of carrier density, which is related to excitation fluence. Diffusion times out of the illuminated spot will also be considered.) Our experimental apparatus allows us to fit multiple lifetimes to a single decay curve, so that if different mechanisms give rise to a collection of lifetimes, we are able to determine them.

The design of our first experiment is shown in Fig. 2. An actively modelocked argon ion laser synchronously pumped a Rhodamine 6G dye laser. After polarization rotation and attenuation, these pulses excited a quantum-well sample. Photoluminescence was passed through a monochromator tuned to the $n = 1 \rightarrow 1$ transition and into a single-photon-counting photomultiplier tube. Time-resolved single-photon-counting techniques were used to determine recombination times. The laser system and single-photon-counting electronics were on loan from the Stanford Structural Biology Department. The best sample available to us for these experiments was a MOCVD single quantum well grown at Xerox PARC by R.D. Burnham. A time-integrated photoluminescence spectrum of this sample is shown in Fig. 3. The photoluminescence at the $n = 1 \rightarrow 1$ transition energy exhibits two decay times, a short one $\sim$2 ns and a long one $\gtrsim$100 ns, as shown in Fig. 4. The prominent long lifetime was not anticipated. After performing this experiment, we learned that the MOCVD reactor in which this sample was grown suffered from a serious silicon contamination problem. This long lifetime could possibly be related to the high silicon concentration in the sample. Another possible explanation is that the long lifetime represents carriers becoming trapped in the barrier region before decaying down into the well. Excitation at a photon energy below the barrier bandgap energy would illuminate this issue.

Our second set of experiments addressed these probems. Since we are attempting to measure an intrinsic lifetime, we are studying an MBE sample grown by A.C. Gossard at AT&T Bell Laboratories. This sample, designed to our specifications, has 100 87Å GaAs wells separated by 194Å $Al_{.26}Ga_{.74}As$ barriers. The larger number of wells means that the effects we observe are certainly dominated by the quantum wells, not the cap or the substrate. The excitation source is a krypton ion laser actively modelocked at 647 nm and 752 nm, on loan from the San Francisco Laser Center. These two wavelengths fall above and below the barrier bandgap enabling us to observe if the carriers are held up in the barrier region. A monochromator

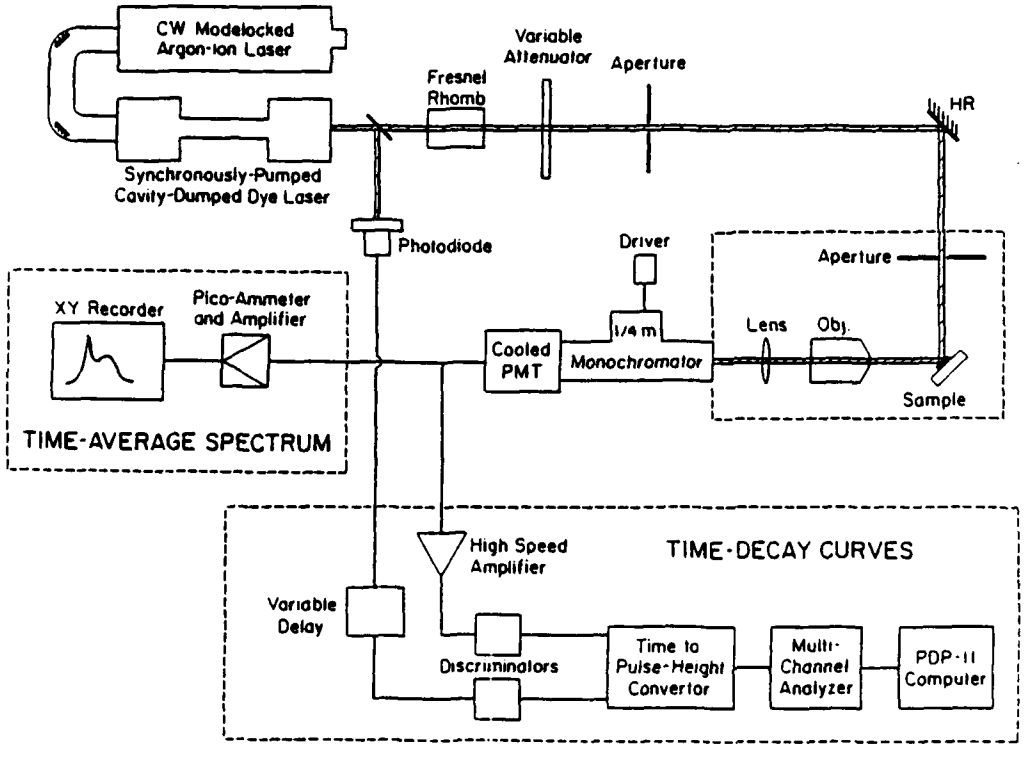

FIGURE 2

Design for Initial Experiments (see text). Dye laser pulse $\sim$10 ps photoexcites carriers in the quantum-well sample. Photoluminescence is collected and filtered before entering a single-photon-counting photomultiplier tube. Amplification of the photomultiplier current, while scanning the monochromator, yields time-integrated spectra. Fast time-resolved single-photon-counting electronics provide decay-time information at a wavelength selected by the monochromator.

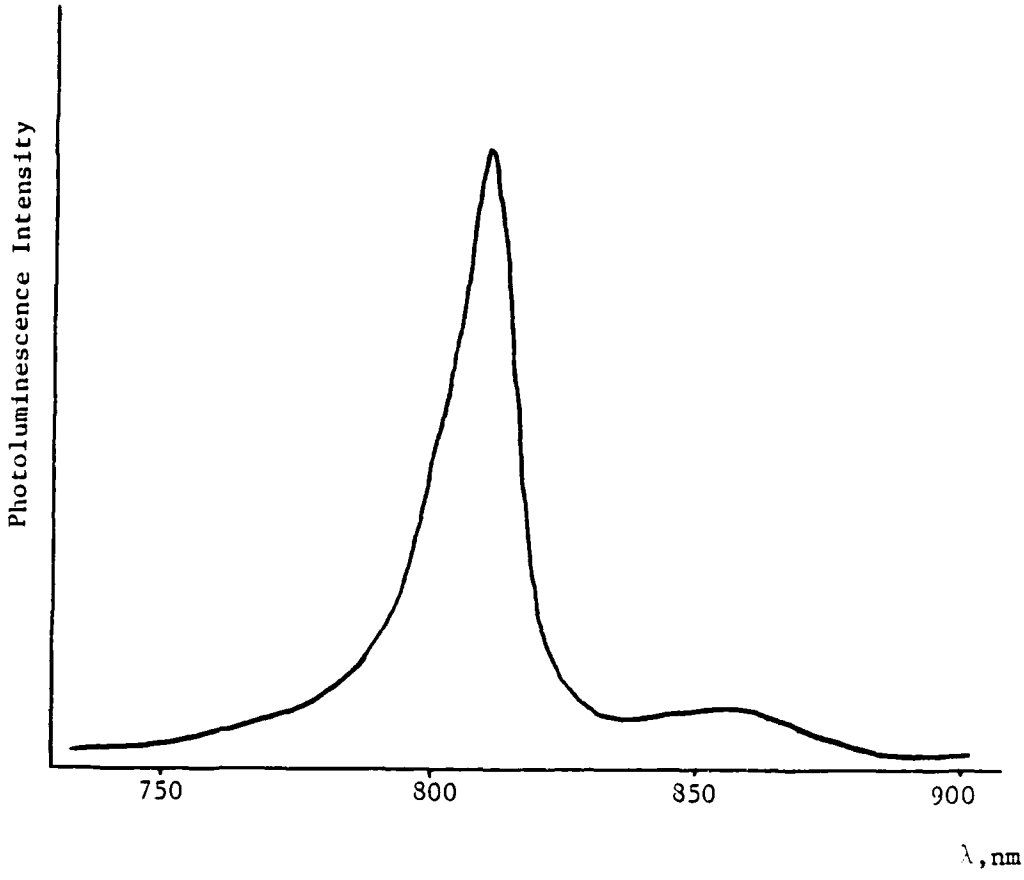

FIGURE 3

Time-Integrated Photoluminescence Spectrum. This spectrum of a single quantum-well grown by MOCVD at Xerox PARC, exhibits two major features. The strongest peak is due to the $n = 1 \rightarrow 1$ heavy-hole transition. The weak peak at longer wavelength is due to the doped thick GaAs cap on this sample.

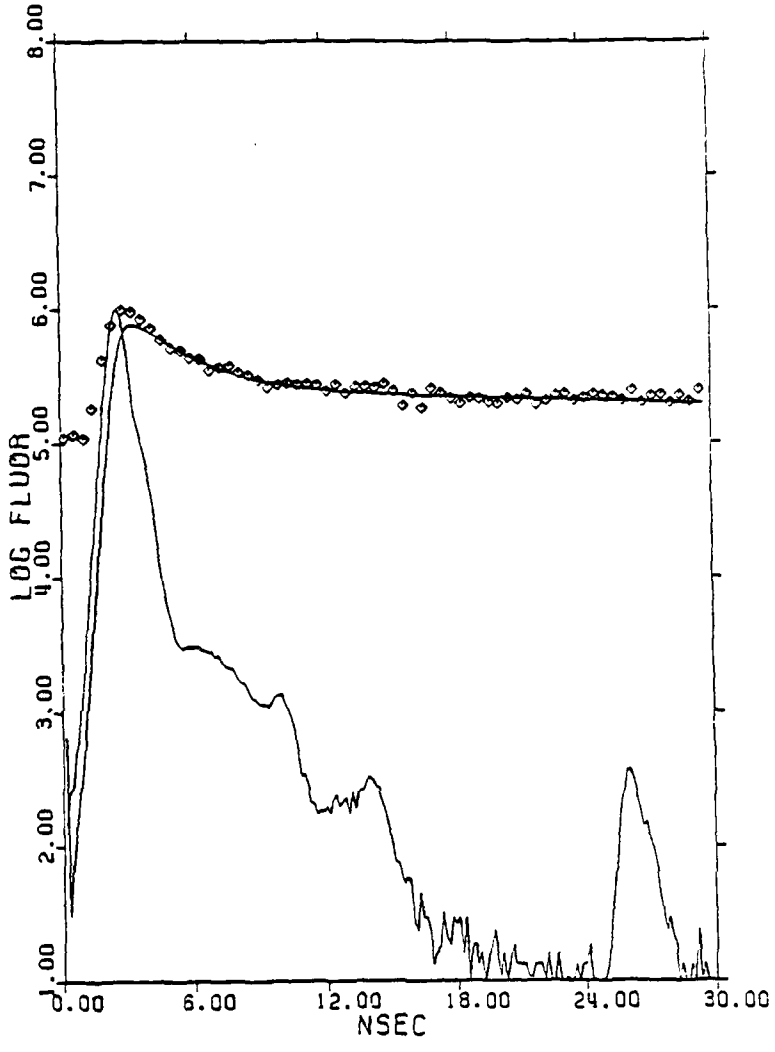

FIGURE 4

Time-Resolved Photoluminsence. This decay curve was taken from the same MOCVD single quantum well at the same excitation energy as Fig. 3. The diamonds represent the photoluminescence when the monochromator is tuned to the $n = 1 \rightarrow 1$ heavy-hole peak. The jagged plot is the impulse response of the detection system at the laser excitation wavelength. The smooth line is a trial fit to the data after deconvolution. The ordinate scale is logarithmic and the full-width of impulse range response is ≈ 1 nsec.

selects the $n = 1 \rightarrow 1$ photoluminescence, and time-resolved single-photon-counting electronics, on loan from the Stanford Physics Department, are used to determine the recombination time. We decided to continue using the time-resolved single-photon-counting technique because its great sensitivity allows us to excite at lower fluences than we would need with other techniques, and it is best to try to measure an intrinsic lifetime at low excitation energy densities. A time-integrated spectrum of the photoluminescence is shown in Fig. 5. A plot of the time decay curve at the $n = 1 \rightarrow 1$ transition wavelength is shown in Fig. 6. This semi-log plot shows three lifetimes: a strong $\lesssim$1 nsec lifetime; a weaker $\sim$3 ns lifetime; and a very weak $\sim$50 nsec lifetime. Current results of this work, which is being carried out by graduate student J.E. Fouquet, have been accepted for presentation at the 1984 International Conference on the Physics of Semiconductors in San Francisco, August 1984.

B. Picosecond Studies on Semiconductors

1. Excite-Probe Experiments

We have performed a series of studies of picosecond optical effects using laser pulses on semiconductors to study picosecond carrier lifetimes, laser annealing, damage and transport effects. One result of this work has been a new technique for observing dense electron-hole plasmas in silicon and gallium arsenide just below melting, by observing surface transformations induced by two variable-delay picosecond pulses at different wavelengths. Using this two-step excitation technique, we find that it is possible to control and "gate" the melting effects of a very intense 1.06 μm pulse by varying the intensity of a much weaker 532 nm "trigger pulse." In essence the 532 nm pulse creates a dense electron-hole plasma, which greatly increases the absorptivity for the 1.06 μm pulse by free-carrier absorption. Varying the pulse intensities and the delay between the 532 nm and 1.06 μm pulses permits us to study the decay of the dense electron-hole plasma in detail.

Our two-step excitation technique has proved to be more sensitive than conventional excite-and-probe experiments at the same wavelengths. In Fig. 7, we show some typical results obtained with our method. By fitting such experimental results to a theoretical model, we have been able to obtain the following parameters in very highly excited ion-implanted silicon: relaxation time = 0.1–1 ps, recombination time = 10 ps. Our results to date are discussed more thoroughly in Refs. [8–11]. We are

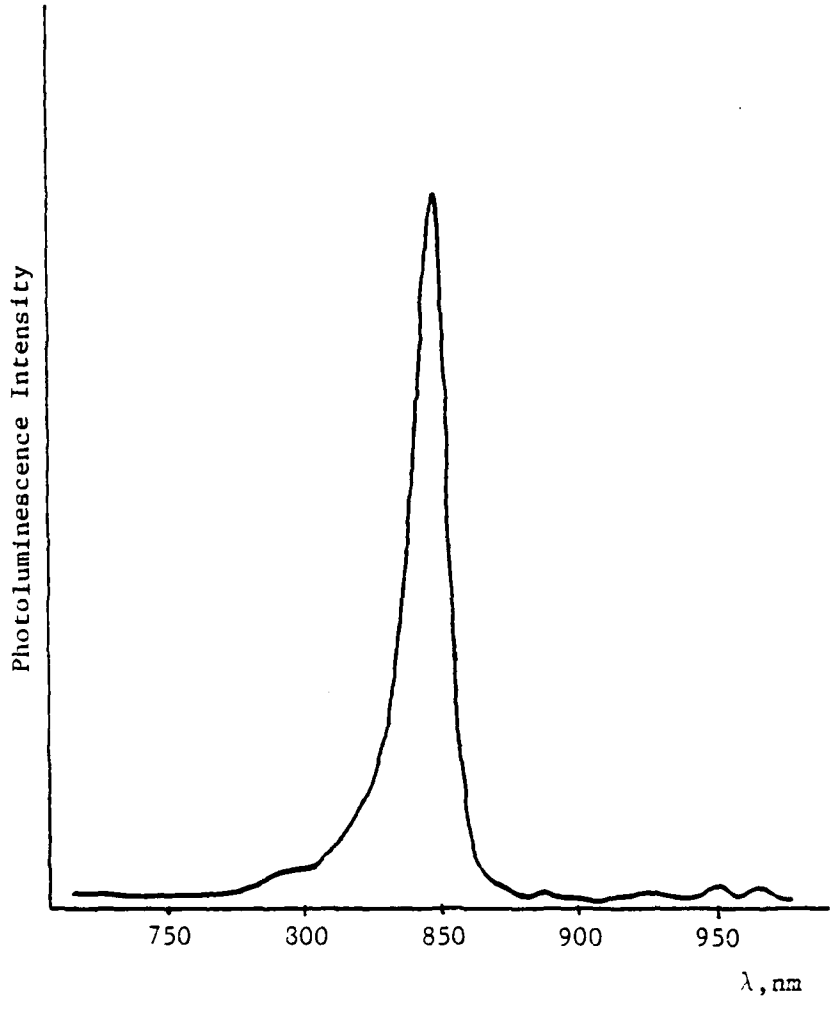

FIGURE 5

Time-Integrated Photoluminescence Spectrum. This spectrum is of an MBE multiple quantum-well structure grown at AT&T Bell Labs. The $n = 1 \rightarrow 1$ heavy-hole transition dominates.

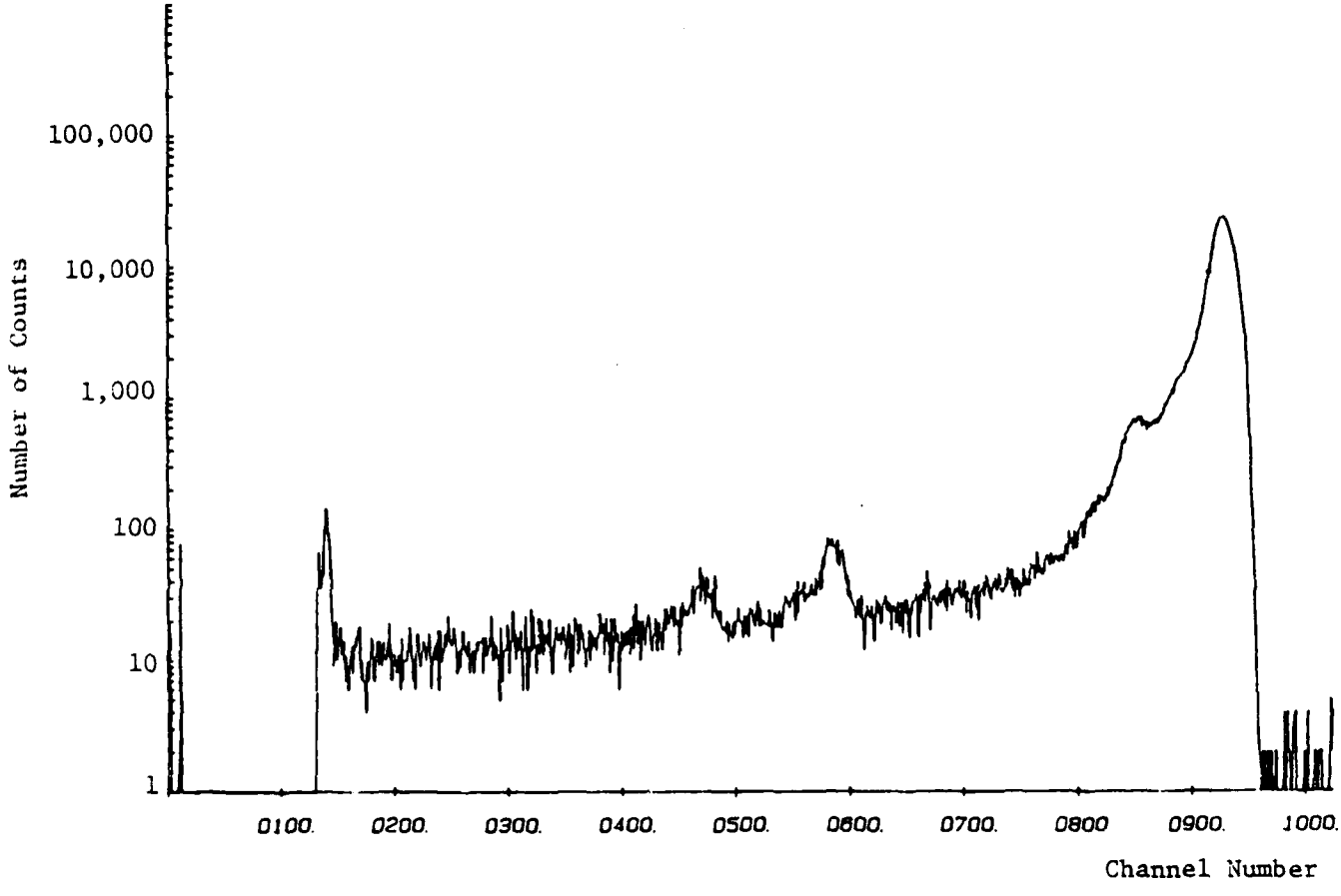

FIGURE 6

Time-Resolved Photoluminescence. This decay curve was taken of $n = 1 \rightarrow 1$ heavy-hole photoluminescence from the same MBE multiple quantum-well sample as Fig. 5. The ordinate scale is logarithmic and the full-width of the impulse response is ≈ 1 nsec. $t = 0$ is at the right-hand side of the plot and time increases to the left. Scale is .12 ns per channel. The two peaks in the center of the plot after $t = 0$ are also present in the impulse response of the system.

continuing these experiments with special emphasis on how the various parameters vary from intrinsic, crystalline to fully amorphized samples.

2. Laser-induced Periodic Surface Structures and Phase Transitions

During laser illumination, spontaneous periodic surface structures or ripples are produced on the surface of practically all solids (metals, semiconductors and insulators). Many laser wavelengths and pulse-lengths have produced these effects. Ripples occur when the light intensity is close to melting threshold for one-pulse illumination or when the intensity is much lower for multiple pulse illumination. We started investigating this puzzling phenomenon when we observed such ripples after laser annealing Si with picosecond pulses. In the past two years, we have made decisive progress in our understanding of the formation of these ripples.

In our own work we have produced ripples on GaAs, Ge, Si and Cu surfaces, with picosecond and nanosecond laser pulses from our Nd:YAG laser system. The well-defined properties of these ripples have been reported in several publications and conferences.[8,13–17] We confirmed some earlier results published on that topic and added new observations. The universal character of that form of damage is directly relevant to the microelectronics industry (laser annealing and processing) and laser-fusion effort (mechanism of surface degration under multiple subthreshold illumination). It is not surprising that many other groups (in the U.S., Canada, Europe and Japan) in past years have also reported experimental results pertaining to surface ripples.

The dependence of the ripple spacing on the angle of incidence and electric field direction suggested that interference between the incident beam and a wave scattered along the surface may lead to a positive feedback situation for ripple growth. We have since then developed these ideas in detail. With our model, which involves solving Maxwell's equation at a periodic interface and computing the Poynting vector flux inside the solid, we have been able to show that a corrugated surface is most favorable for ripple growth while a periodic variation of the electron-hole plasma density does not produce ripples. Our results have been published in a theoretical paper that appeared in *Physical Review B*.[14] Our model is the first detailed theory that describes the production and growth of spontaneous periodic surface structures. in another publication,[8] we have also used our model to explain

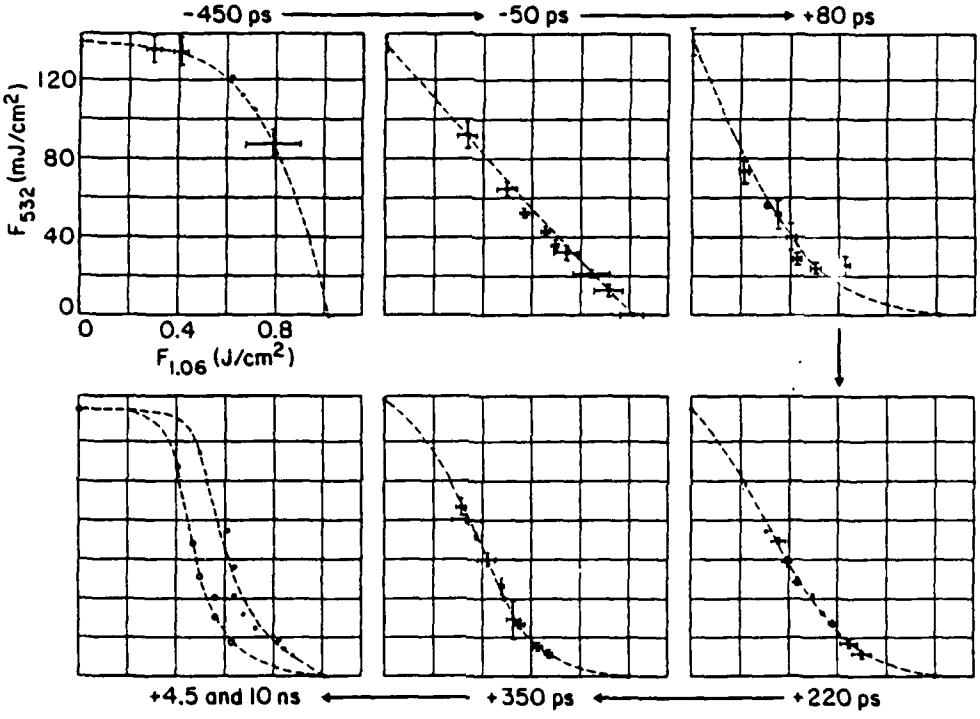

FIGURE 7

Observed pulse-energy melting thresholds on As-implanted Si for joint illumination by $\sim$40 ps pulses at 532 nm and 1.06 μm for various relative delays of the IR pulse.

how strong ripples may occur when the surface is not homogeneously molten.

Solid surfaces may also become rippled after illumination by multiple laser shots below the threshold for melting. In fact, it seems that all surface transitions (crystalline to amorphous, amorphous to crystalline, damage, etc.) may be gradually produced with multiple (picosecond) laser pulses. Our experimental results have recently been published;[18] they show that each gradual transition follows a "universal" growth curve (Fig. 8). An important consequence of this work is that measuring the single-shot damage threshold is almost meaningless, since repetitive illumination with many subthreshold laser pulses may produce surface melting. The true "safe level" is usually a factor of three below single-shot threshold for crystalline materials, and a factor of ten below single-shot threshold for amorphous materials.

We have solved the electromagnetic part of the ripple's formation problem but the material's behavior is not well understood. For example, how can the surface be deformed during a period of time as short as a few picoseconds, and what are the material-science implications of this determination? We are also searching for a model to explain the multiple-shot results and new experimental techniques more sensitive to microscopic transformations. We are presently working on these and related problems. In one new technique we monitor the diffraction pattern of a probe beam as a function of ripple growth. Figure 9 shows some recently published results[15] obtained using this technique. Some of the observations contained in that paper cannot be explained within the framework of one theoretical model (or that of others, for that matter). We have developed a phenomenological interpretation which we are trying to reconcile with our previous understanding. In another theoretical investigation we are trying to estimate the effect of cumulative plastic deformations in our multiple-shot results. In general we shall continue to investigate the various phenomena that occur during pulsed laser illumination of solid surfaces.

C. Tunable-Laser-Induced-Grating Spectroscopy

One of the major experimental activities under this contract has been the development of a promising new technique for using tunable-laser-induced material gratings to measure subpicosecond physical processes and relaxation rates. This new "frequency-domain" technique should have a time resolution beyond the reach

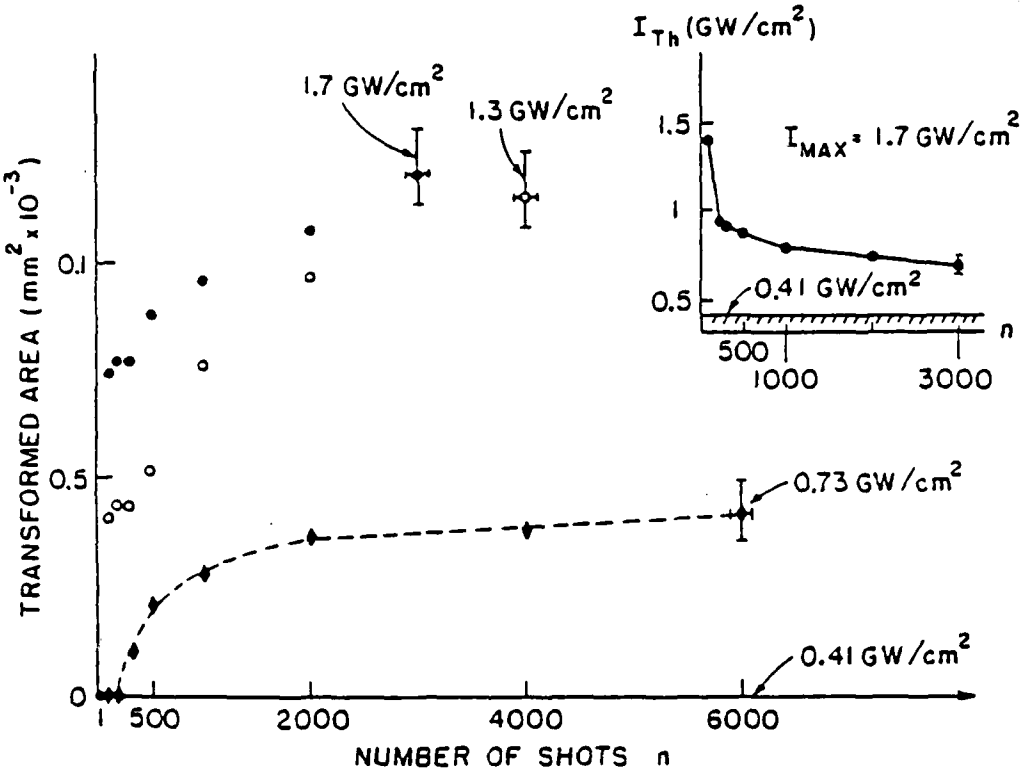

FIGURE 8

Transformed area versus number of shots in crystalline Si illuminated by 80 ps, 532 nm laser pulses. The inset shows the decrease in threshold for $I_{MAX} = 1.7$ GW/cm^2. The threshold for the three different intensities approaches 0.41 GW/cm^2 as n gets large. The repetition rate of the laser is 100 Hz.

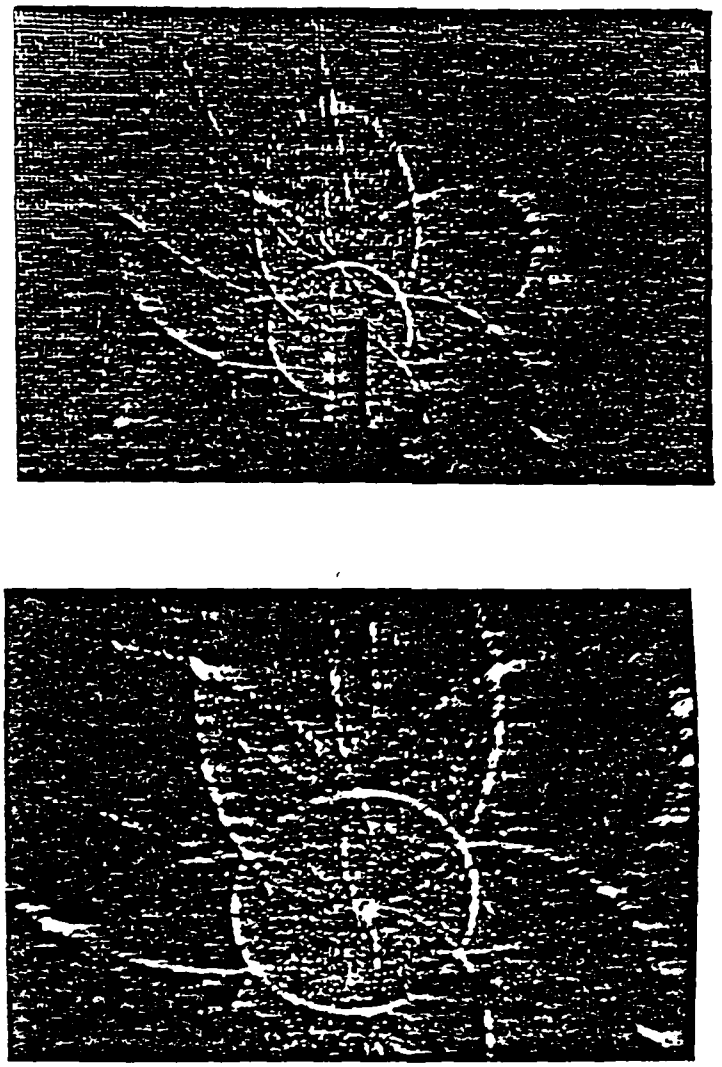

FIGURE 9

Diffraction pattern of rippled (111)Ge surfaces after illumination with multiple circularly polarized 106 μm laser pulses at normal incident. The probe beam is a cw A_2^+ laser

of any time-domain ultrashort-pulse method. This work has been under the primary direction of Rick Trebino, a graduate student who has continued this research after graduation as a research associate.

The approach in this experiment has been to generate in an experimental sample a moving laser-induced grating of excited states created by the interference fringes between "excitation beams" from two tunable lasers with slightly different frequencies, and then to observe the diffraction from this moving grating with a third "probe beam" at another wavelength. Measurement of the diffracted intensity as a function of excitation-beam frequency difference results in the measurement of ultrafast relaxation times of the excited states created by the excitation beams.

The experimental apparatus required to perform Tunable-Laser-Induced-Grating experiments, consisting of a frequency-doubled Q-switched Nd:YAG laser and three tunable dye lasers, reached completion prior to this contract period. In constructing this apparatus, we found it useful to evaluate the performance of several popular dye-laser designs in order to achieve the performance required for our experiments. The publication resulting from this evaluation[20] compared the Hänsch,[21] Cassegrain,[22] prism,[23] and grazing-incidence[24] designs, which employ a refractive-lens, a Cassegrain-telescope, a four-prism, and a grazing-incidence-diffraction-grating beam expander, respectively. We concluded that the best performance can be had with the prism or grazing-incidence designs, depending on the desired linewidth. The Cassegrain design is similar in many ways to the Hänsch design, each yielding (unnecessary) two-dimensional beam expansion. The Cassegrain design, however, suffers from several problems unique to it, including a diffraction-grating damage mechanism. We concluded that, for our purposes, three grazing-incidence dye lasers would prove optimal.

With these dye lasers, we have successfully performed tunable-laser-induced-grating experiments using a simple beam geometry employing only two lasers. This geometry, introduced by Song, Lee, and Levenson,[25] combines one excitation beam and the probe beam into a single beam (provided by the same laser) with each propagating as a different polarization. With this arrangement, we have measured ultrashort lifetimes of the organic dye, malachite green in water ($\sim$.6 psec),[26,27] and the nonlinear-refractive-index material, carbon disulfide ($\sim$2.5 psec).

Various drawbacks inherent in this two-laser geometry limited the quality of

these experiments: (1) a significant background, due to an induced-dichroism (or induced-birefringence) effect, severely limited the experimental signal-to-noise ratio; (2) the use of crossed-polarizers provided another background due to polarizer leakage, which severely distorted weak experimental lineshapes; and (3) the equality of probe and excitation wavelengths resulted in the simultaneous probing of both a ground-state absorption grating and an excited-state stimulated-emission grating and introduced undesired ambiguities into the experiment. As a result we developed, and obtained excellent results with, a three-laser geometry which is background-free and very versatile, allowing the experimenter free reign in his or her choice of probe wavelength. We also developed a simple alignment procedure for such experiments. Experiments on malachite green in water reported in the *Journal of Chemical Physics*,[28] and also at conference talks[26,29] (see Fig. 10), yielded much less noisy data in one tenth of the experimental time required in our previous two-laser experiments and revealed a non-Lorentzian lineshape. This lineshape proved inconsistent with a simple exponential decay but agreed quite well with a two-component exponential (.78 and 7.4 psec), and also a theoretical decay resulting from the presence of a range of lifetimes (from .34 to 7.2 psec). Previous (time-domain) experiments by others have lacked the temporal resolution to see such complex ultrafast behavior.[30]

We have also studied the problem of the simultaneous formation of thermal gratings with excited-state gratings resulting in the obscuration of the latter. We developed a mathematical theory of the formation of thermal gratings which necessarily involved the partial-coherence properties of the excitation lasers. Involving fourth-order coherence functions, this theory predicts the existence of grating fringes under many circumstances when a standard second-order theory (used to describe the Michelson interferometer, for example), which is not appropriate for this problem, does not. In particular, this theory predicts the existence of thermal gratings in tunable-laser-induced-grating experiments, while the second-order theory does not.

The thermal-grating theory can also be applied to experiments in which two beams (one delayed) from a single laser induce the grating. Under many circumstances, the expected diffraction efficiency as a function of delay of an induced thermal grating will be the sum of two terms: (1) a "spike" with a width cor-

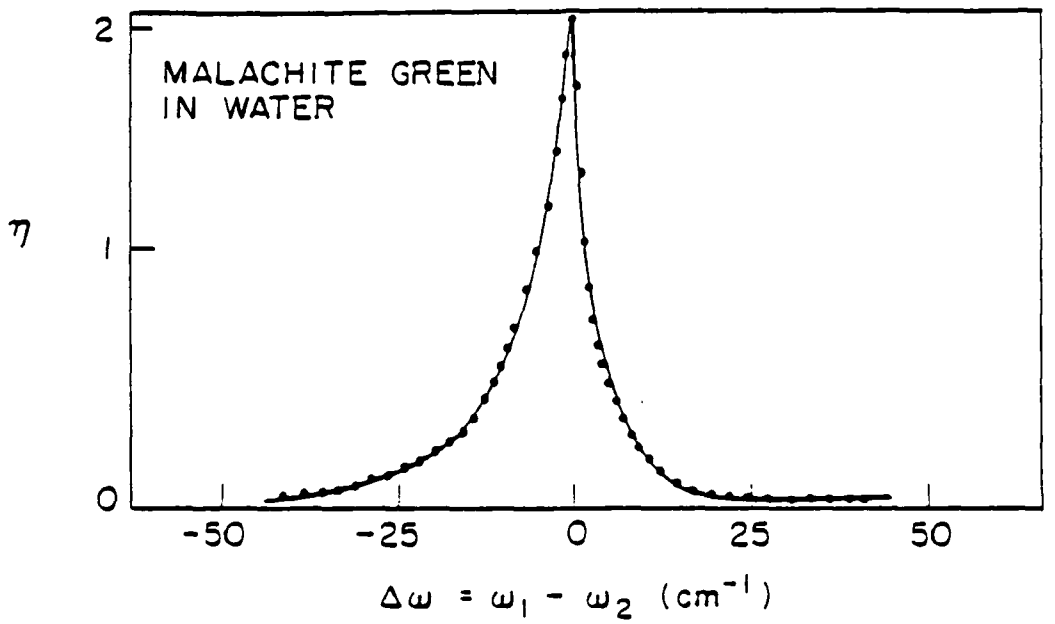

FIGURE 10

Three-laser tunable-laser-induced-grating experiment on malachite green in water. The solid line represents the theoretical lineshape corresponding to decay curves consisting of both the sum of two exponentials and a uniform distribution of exponential decays. See text for numerical results. (Vertical-scale units are arbitrary).

responding to the radiation coherence time, and (2) a broader peak corresponding to the pulse autocorrelation function. As a result, a thermal-grating experiment can yield both the coherence time and pulselength of the excitation radiation. Figure 11, showing data from an experiment previously performed by Eichler, Klein, and Langhans,[31] illustrates this observation quite well. The dashed line illustrates the second-order result, fit by Eichler *et al.*, while the solid line shows the best fit of our fourth-order theory, which agrees with the independently measured pulselength. Since automatically phasematched, background-free geometries exist for the performance of thermal-grating experiments, and thermal-grating effects are quite strong, such a pulselength measurement scheme may be preferred to conventional SHG techniques.[26]

D. Phase Conjugation and Phase Conjugate Resonators

During the contract period a number of theoretical projects related to optical phase conjugation and phase conjugate resonators were carried out and published.

As one major part of this, we have carried out over the past two years a detailed theoretical analysis of phase conjugate resonators or "PCM resonators," that is, optical resonators in which one or both mirrors are optical-phase-conjugation elements. The results of this analysis, besides appearing in journal publications, have been collected in a lengthy chapter in the book, *Optical Phase Conjugation* edited by Fisher[32] published in 1983.

Phase-conjugate resonators have been found to have novel and useful features in both their transverse and axial modes. For instance, the transverse modes in a PCM resonator are always "stable," i.e., there is always a perturbation-stable and confined gaussian mode, no matter how divergent or unstable the conventional end mirror curvature may be. Figure 12 shows for example the spot sizes and mode profiles inside a PCM resonator with one conventional mirror and one soft-apertured phase conjugate mirror, as a function of the curvature or the g parameter for the conventional mirror.

The axial modes of phase conjugate resonators also differ from conventional cavities, occurring as symmetric pairs of "half-axial modes," i.e., the resonant modes are spaced by $c/4L$ Hz rather than the usual $c/2L$ Hz, where L is the cavity length. Figure 13 shows graphically how an off-resonance signal traveling inside

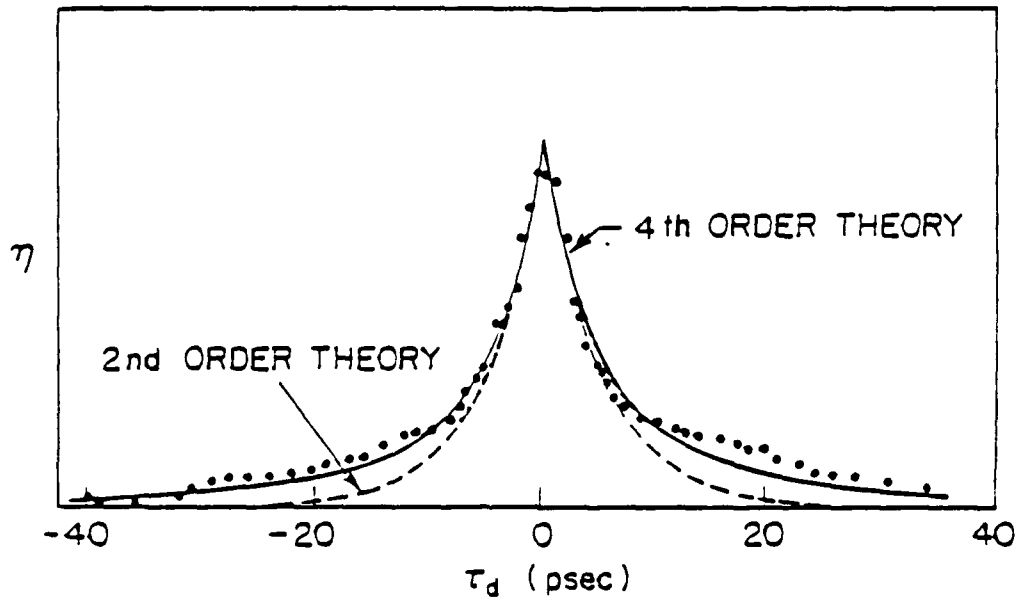

FIGURE 11

Thermal-grating diffraction efficiency vs. relative delay between excitation beams. Dashed line shows the best fit of the second-order theory used by Eichler *et al.* The solid line shows the best fit of the fourth-order theory, yielding a value for the laser pulselength in addition to the coherence time.

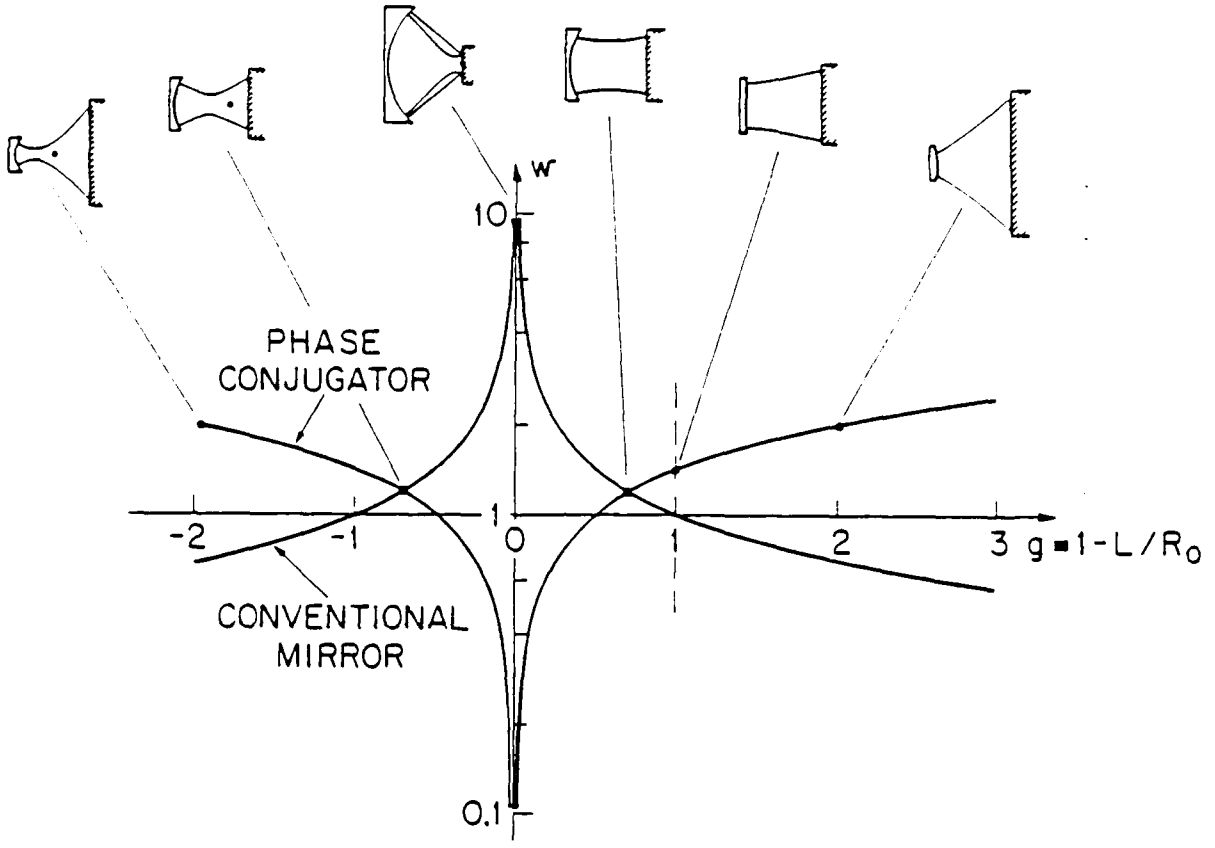

FIGURE 12

Gaussian spot sizes at the conventional-mirror and phase-conjugate-mirror ends of an elementary PCM resonator, plotted versus the resonator parameter $g \equiv 1 - L/R_0$, where R_0 is the curvature of the conventional mirror. The resonator is assumed to have a weak gaussian aperture just in front of the conjugator; but the spot sizes are independent of the actual size of this aperture.

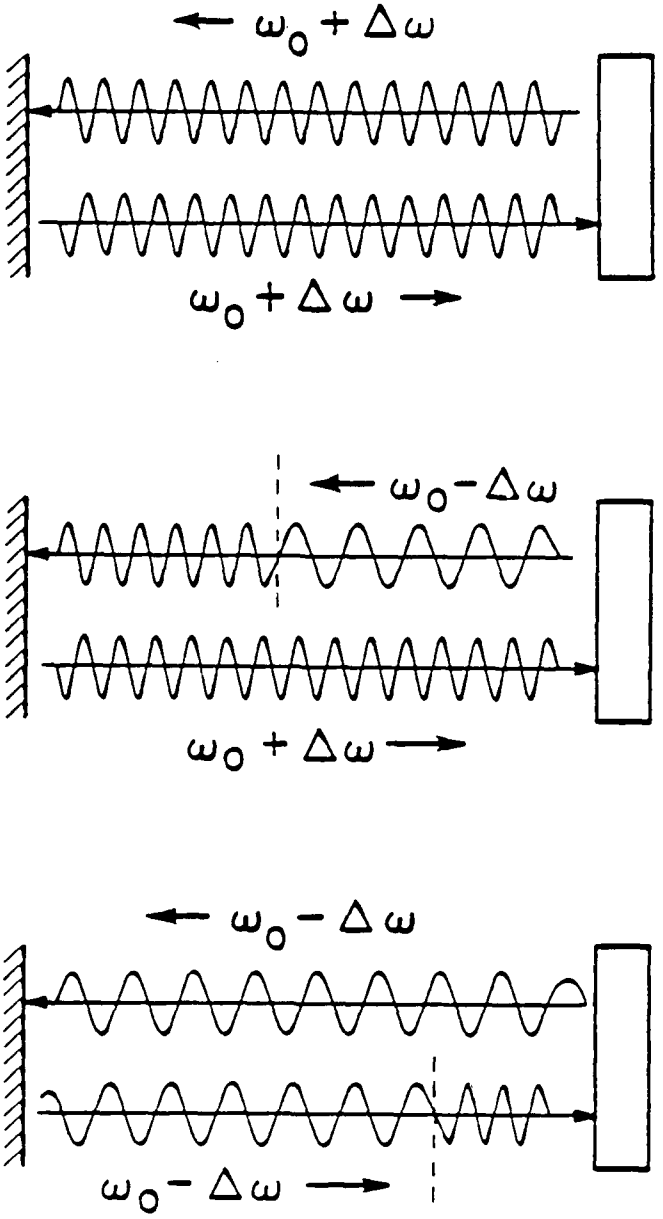

FIGURE 13

A wave circulating inside a PCM resonator at an off-resonance frequency $\omega_0 + \Delta\omega$ will all be converted to the opposite value $\omega_0 - \Delta\omega$ after one round trip, and then back again after another round trip. Hence it takes two full round trips for the initial wave to be fully reproduced, and the axial mode spacing for the cavity is $c/4L$ instead of $c/2L$.

a PCM cavity will be shifted from above to below resonance on one round trip, coming back again on the next round trip; so that it takes *two* round trips for the signal to reproduce completely, and hence the axial modes are spaced by *half* the usual $c/2L$ value. (Our prediction of this behavior has recently been confirmed by experimenters at the Hughes Research Labs).

If phase-conjugate resonators are to be used as oscillators, one needs to know the large-signal behavior of such devices, including pump depletion, nonlinear refractive index changes (optical Kerr effect), signal saturation and distributed losses. With all these effects included, the PCM resonator is described by four complex nonlinear differential equations for which no analytic solutions exist. We have therefore developed a simple iterative numerical procedure for finding solutions to these equations. This approach, and the results of such calculations for various cases of interest, have been reported in a recently published paper.[33] Figure 14 shows for example the normalized phase-conjugate power output versus output mirror reflectivity from a phase conjugate resonator which contains a laser gain medium with different unsaturated gain coefficients g_0L. One important conclusion from these calculations is that the larger fraction of the power output from such a cavity comes from the phase conjugate mirror end, coming through the phase conjugator, and not from the conventional mirror end; and this energy is *not* phase conjugated, so that intracavity phase distortions are *not* removed.

The basic orthogonality properties of PCM resonator modes were also worked out and published,[34] and two invited conference presentations on phase conjugate resonators were given.[35,36]

E. Picosecond Photoacoustic Spectroscopy

During this period, in work supported in part also by NASA, we successfully demonstrated that unique picosecond measurements can also be made by combining picosecond optical pulses with photoacoustic detection. Photoacoustic detection and photoacoustic spectroscopy (detection of light-induced sound) are extremely useful for measuring very weak optical absorptions, or absorptions in nonfluorescing atom and molecular systems. By using a unique elliptical acoustic cell that we invented, and measuring the total acoustic impulse as a function of delay between two picosecond pulses at different wavelengths, we are able to observe very weak

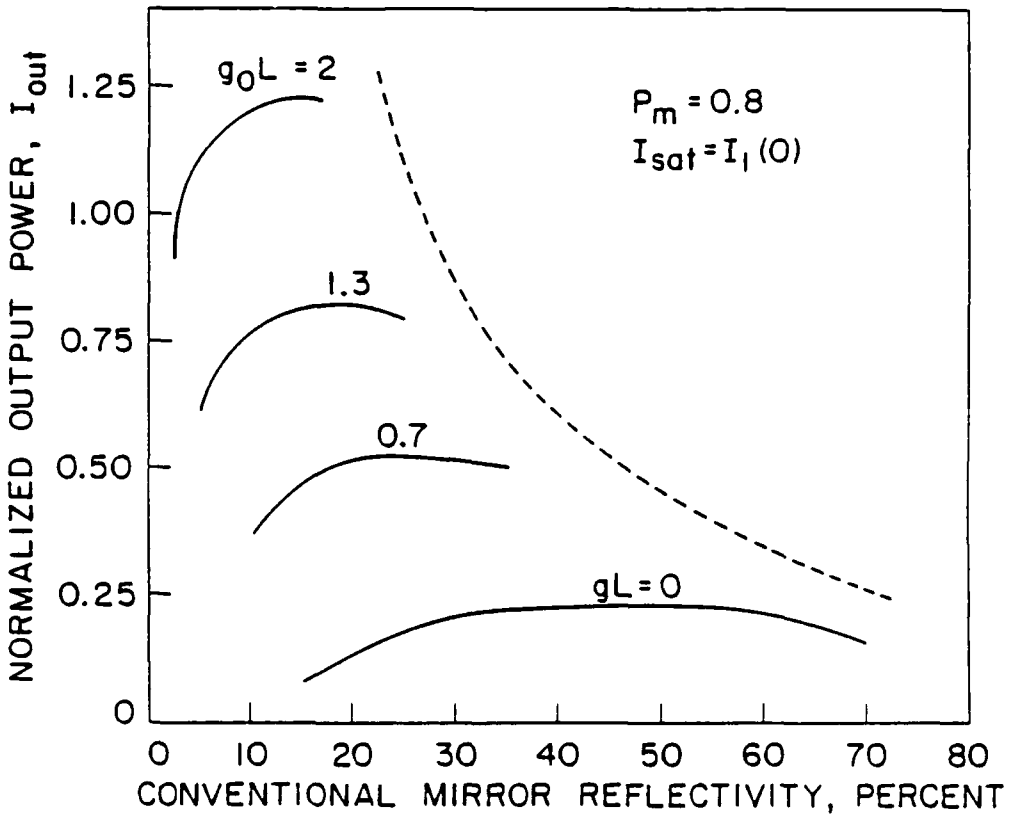

FIGURE 14

Power output from a phase conjugate oscillator, normalized to the total pumping power of a DFWM cell, plotted versus the reflectivity of the conventional mirror, for different values of the unsaturated gain coefficient of a laser medium placed inside the cell. The saturation intensity of the laser medium is assumed equal to the one-way threshold pump power for the DFWM cell.

excited-state absorptions, and observe the excited state lifetime with picosecond time resolution.[37–39] Figure 15 illustrates typical results from this technique for an organic dye solution. We believe this technique will provide a very useful melding of the capabilities of picosecond and photoacoustic spectroscopy.

F. Colliding-Pulse Mode Locking

The most important development in laser mode-locking during the past few years has been the colliding-pulse saturable-absorber technique in cw-pumped dye lasers by Shank and co-workers at AT&T Bell Laboratories. Following this discovery, we proposed a novel "antiresonant ring" (ARR) cavity for achieving colliding-pulse mode-locking in solid-state lasers such as Nd:YAG. Experiments carried out by collaborators at the Free University of Brussels then seemed to verify the advantages of this approach with measured pulsewidths as short as 12 to 15 psec. These and other results are described in two invited review papers[40,41] prepared during the reporting period.

During the reporting period, we assembled a Nd:YAG laser of similar design and performed an extensive series of antiresonant-ring colliding-pulse experiments. Figure 16 shows, for example, an autocorrelation trace of the mode-locked laser pulse. We found the ARR cavity to be easy to align and mode-locking easy to obtain. The mode-locked laser is considerably more stable than the standard laser cavity with a cell containing the mode-locking dye contacted to the rear mirror. This stable behavior is most likely due to the absence of reflections from the dye cell coupling back into the laser in our cavity; the contacted dye cell of the linear cavity reflects a small portion of the light back into the direction of the laser, which can disturb mode-locking.

We were, however, not able to reproduce the extremely short pulsewidths obtained in the Belgian experiments. Over a wide range of cavity dimensions, ratios of spot size in the Nd:YAG rod to that in the saturable-absorber dye cell, pumping energy density for the Nd:YAG rod, dye concentrations (with Eastman Kodak 9860 and 9740 dyes), and a host of other parameters, we always obtained pulses in the usual Nd:YAG range of 25 to 30 psec. The only parameter which yielded significantly different pulsewidths was moving the dye cell from the center of the antiresonant ring. In this case, the laser would not mode-lock as well and the

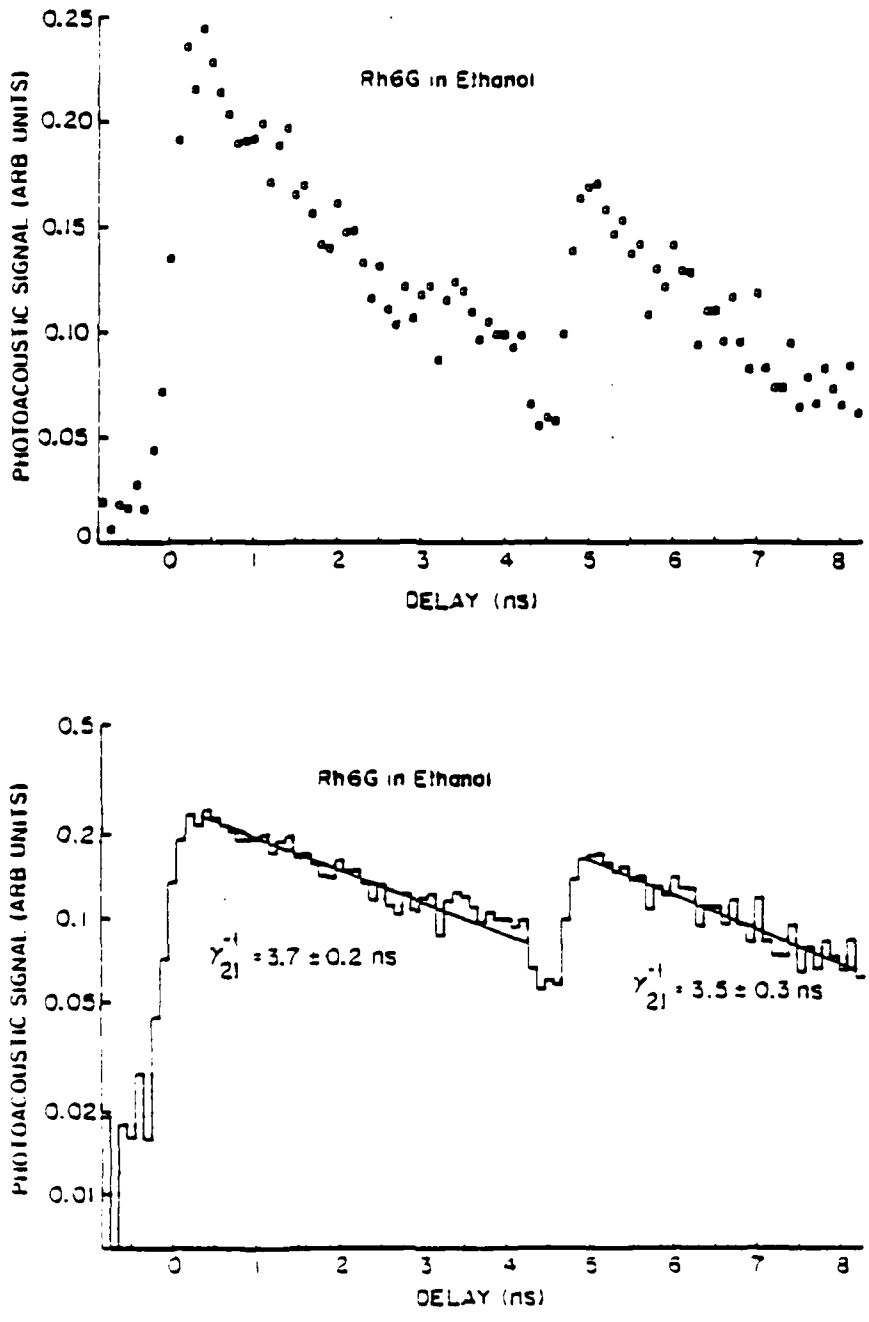

FIGURE 15

Picosecond time-resolved photoacoustic results for Rhodamine 6G in ethanol. Top trace: linear scale. Bottom trace: log scale and best fit to experimental data.

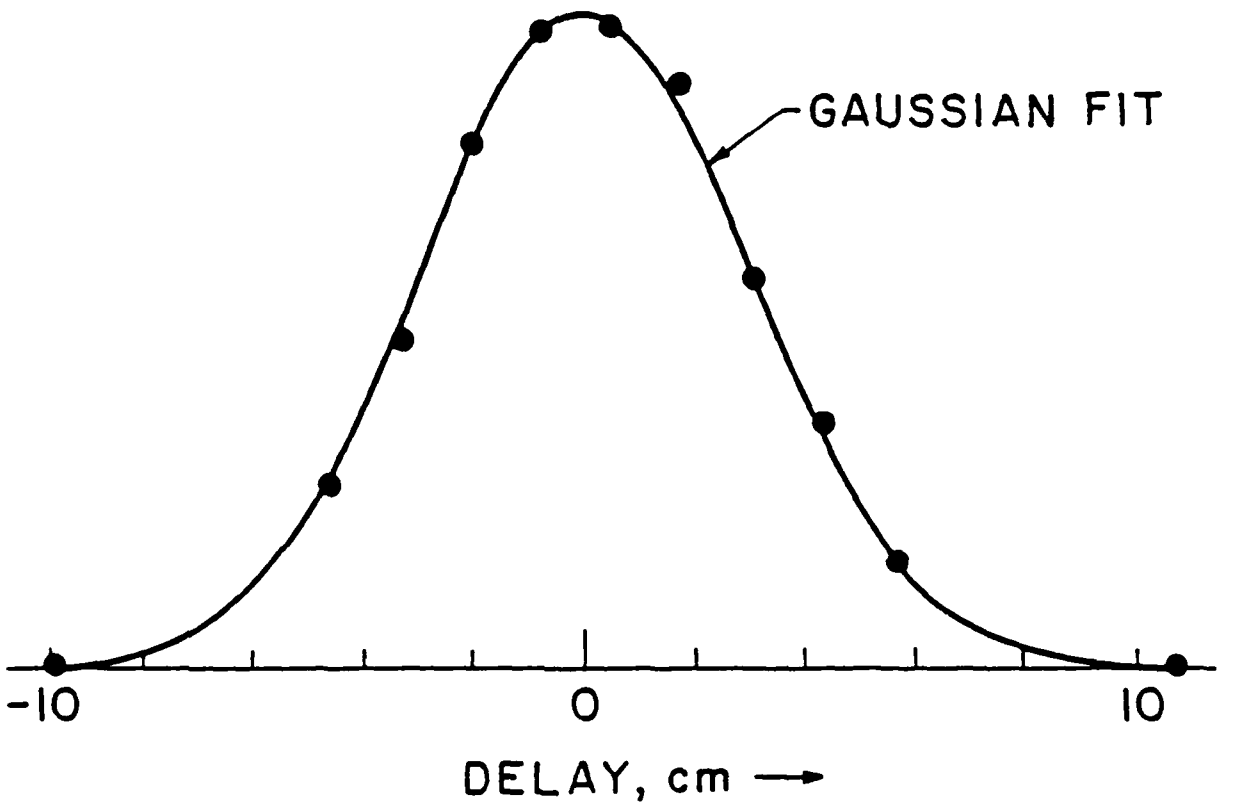

FIGURE 16

Background-free SHG autocorrelation trace for the mode-locked YAG laser using an antiresonant ring cavity.

pulsewidth would lengthen.

Our pulsewidth measurements were made using background-free SHG autocorrelation which averaged the pulsewidth over all pulses in the Q-switched train and over many trains. The Belgian measurements used two-photon fluorescence and (very briefly) a streak camera. Two-photon fluorescence is not a background-free measurement, which leads to some uncertainties in determining the pulsewidth. It does, however, measure the widths of the pulses in individual Q-switched trains. The Belgian experiments obtained the short 12 to 15 psec pulses only on certain occasions. Passive mode-locking is very complicated and is well-documented to be subject to wide statistical variations in output pulse energy and pulsewidths. It is conceivable that our laser and the Belgian laser both sometimes produce pulses as short as 15 psec. Our measurements state only that the minimum pulsewidth averaged over many Q-switched trains in passively mode locked Nd:YAG is limited to roughly 30 psec. We always obtained similar results over a wide range of cavity parameters (our experiments were far more extensive than those leading to the Belgian group's publications), pointing to a fundamental limit in Nd:YAG.

Back-of-the-envelope calculations indicate that dispersion in the Nd:YAG rod due to the variation in refractive index of Nd:YAG across the linewidth of the 1.064 μm transition could be responsible for broadening the pulsewidth beyond its Fourier-transform limit and lead to $\approx$30 psec pulses. Furthermore, currently accepted colliding-pulse mode-locking theory predicts a significant dependence of pulsewidth on the ratio of beam size in the Nd:YAG rod to beam size in the saturable-absorber dye cell. We tried a wide range of beam area ratios both greater than and less than 1, finding the pulsewidth to be insensitive to this ratio. If the theory is at all correct, these results indicate that some other factor is broadening the pulses in a Nd:YAG laser, and dispersion in the Nd:YAG rod is the most likely cause.

We feel the results are best explained by a combination of both of the above-mentioned effects: that is, dispersion in the Nd:YAG rod is limiting the average pulsewidth to 30 psec, but statistical variations in the mode-locking process (such as: at what energies the gain narrows and the absorber saturates) yield a wide range of pulsewidths, some of which are as short as 15 psec.

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III. AFOSR-SUPPORTED PUBLICATIONS AND CONFERENCE PAPERS

1. A. Hardy, P.A. Belanger and A. E. Siegman, "Orthogonality Properties of Phase Conjugate Optical Resonators," *Appl. Opt.* **21**, 1122–1124 (15 March 1982).
2. P.M. Fauchet and A.E. Siegman, "Surface Ripples on Silicon and Gallium Arsenide under Picosecond Laser Illumination," *Appl. Phys. Lett.* **40**, 824–826 (1 May 1982).
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21. A.E. Siegman, "Optical Device Applications for Photoactive Solids," (invited), presented at Emil Warburg Symposium on Unconventional Photoactive Solids, Wasserschloss Mitwitz, Germany, June 1982.
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24. J.M. Heritier and A.E. Siegman, "Picosecond Time-Resolved Measurements Using Photoacoustic Detection," presented at Third International Topical Meeting on Photoacoustic and Photothermal Spectroscopy, Paris, March 1983.
25. A.E. Siegman, "Principles of Lasers (series of 15 lectures)," presented at Far Eastern Laser School (FELS–83), Seoul, Korea, July 1983.
26. J.M. Heritier and A.E. Siegman, "Time-Resolved Excited State Measurements Using Photoacoustic Detection," presented at 4th International Conference on Dynamical Processes in Excited States of Solids (DPC-83), Stanford, California, July 1983.
27. P.M. Fauchet and A.E. Siegman, "Multiple Pulse Effects in Laser-Induced Transformations of Semiconductor Surfaces," presented at Americal Physical Society Meeting, San Francisco, California, November 1983.
28. A.E. Siegman, "On the Fringes: Laser-Induced Grating Effects," (invited), presented at *LASERS–83*, San Francisco, December 1983.

IV. PROFESSIONAL PERSONNEL

Professor A.E. Siegman continues as Principal Investigator of this contract.

Graduate students associated with this contract at various times during this period were Charles Barker, Julie Fouquet, Philippe Fauchet, Jean-Marc Heritier, Rick Trebino, David Ress, Karen Liu, and Nancy Missert.

Also taking part in the research activities during this period have been Dr. Yao Jian-quan of Tianjin University, People's Republic of China, Mr. Zhou Guosheng, of Shanxi University, People's Republic of China, Dr. Kenju Otsuka of NTT Laboratories, Japan, and Dr. Celso P. Ladera of Universidad Simon Bolivar, Caracas, Venezuela.

Ph.D. dissertations completed during the period of this report include:

Jean-Marc Heritier, *Picosecond Spectroscopy Using a Photoacoustic Detector*, March 1983.

Rick Trebino, *Subpicosecond Relaxation Studies Using Tunable-Laser-Induced Grating Techniques*, May 1983.

Philippe M. Fauchet, *Picosecond Laser Induced Surface Transformations in Semiconductors*, October 1983.

V. HONORS, AWARDS, AND OUTSIDE INTERACTIONS

Several invited conference presentations were based primarily on work done under this AFOSR support, as listed in Section III of this report, including invited papers at the 1982 OSA Topical Meeting on Picosecond Phenomena, the 1982 NATO Symposium on the Application of Picosecond Lasers in Chemistry, the Warburg Symposium on Unconventional Photoactive Solids, the 1982 International Workshop on Phase Conjugation and Instabilities, the 13th Winter Colloquium on Quantum Electronics, and at LASERS-83.

Numerous government and industrial personnel have visited the laboratory, in addition to members of the Solid-State Affiliates Program.

Professor Siegman has continued as an active member of the Air Force Scientific Advisory Board, including Chairman of the Basic Science Panel (formerly the Panel on Research and Geophysics). Dr. Philippe Fauchet has been the recipient of an IBM post-doctoral fellowship, and was the Student Award Winner of the 1983 Material Research Society Annual Meeting; and Julie Fouquet has received the Zonta International Amelia Earhart Fellowship.

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